

# Complex oscillatory yielding of model hard sphere glasses

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## Abstract

The yielding behaviour of hard sphere glasses under large amplitude oscillatory shear has been studied by probing the interplay of Brownian motion and shear-induced diffusion at varying oscillation frequencies. Stress, structure and dynamics are followed by experimental rheology and Brownian Dynamics simulations. Brownian motion assisted cage escape dominates at low frequencies while escape through shear-induced collisions at high ones, both related with a yielding peak in  $G''$ . At intermediate frequencies a novel, for HS glasses, double peak in  $G''$  is revealed reflecting both mechanisms. At high frequencies and strain amplitudes a persistent structural anisotropy causes a stress drop within the cycle after strain reversal, while higher stress harmonics are minimized at certain strain amplitudes indicating an apparent harmonic response.

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Hard Sphere (HS) colloids have been used as model systems to study a plethora of fundamental condensed matter problems such as the interplay between equilibrium phases (crystals and liquids) and non-ergodic states (glasses or gels), and their behavior under external fields such as shear[1, 2]. A major goal is to develop an understanding that will enable tailoring of the mechanical and flow properties based on the structure and dynamics at the particle level. HSs form metastable glasses above a volume fraction of about  $\varphi_g \sim 0.59$  where crystallization and long-time diffusion are suppressed[3, 4]. Such states exhibit solid-like behavior at low stresses/strains and shear-melting (yielding) above a yield stress/strain[5]. This phenomenon is investigated more effectively with a combination of rheological and optical or scattering techniques that may unravel the link between microstructure, dynamics and mechanical properties in steady[6, 7] or oscillatory shear [8, 9]. Such studies have shown that particle cages are deformed under shear and particles move irreversibly when a critical strain is exceeded, leading to flow.

Experimentally, large amplitude oscillatory shear tests are widely used to monitor yielding due to their simplicity and relation with linear elastic and viscous moduli,  $G'$  and  $G''$ . However, their interpretation in the non-linear regime becomes complex due to stress distortion introducing higher harmonics[10]. In a wide range of systems (emulsions, polymers and colloids) a generic peak in  $G''_1$  (viscous modulus of the fundamental frequency) is observed representing an increased energy dissipation near the yield strain,  $\gamma_y$ , where  $G'_1 = G''_1$ , beyond which the sample flows[9, 11]. On the theory side, Mode Coupling Theory (MCT)[12] as well as the semi-phenomenological Soft Glassy Rheology (SGR) [13] are able to capture some aspects of Large Amplitude Oscillatory Shear (LAOS) such as the peak of  $G''_1$ . Nevertheless, the underlying mechanisms relating stress with shear induced structure such as cage deformation, breaking and reformation, as well as particle displacements, are poorly understood, and the frequency dependence is largely unexplored.

LAOS experiments are expected to provide valuable information on the interplay of Brownian motion and shear during yielding of HS glasses. Along this line light scattering-echo experiments probing the average particle displacements under oscillatory shear[8, 9] revealed a transition to irreversibility beyond a critical strain amplitude[8, 9]. This can be viewed as the analogue of the shear induced irreversibility observed in concentrated non-Brownian particles[14], since in colloidal glasses out-of-cage diffusion is frozen, while in non-Brownian particles diffusion is absent at all length scales. While experiments combining rheometry

with scattering or microscopy are rather demanding [8, 15–18], computer simulations can provide an alternative route.

Here we use a combination of oscillatory shear rheometry and Brownian Dynamics (BD) simulations to investigate the links between structure and particle dynamics with the non-linear rheological response of HS glasses in a wide range of frequencies,  $\omega$ , non-dimensionalized by  $Pe_\omega^0 = \omega\tau_B$ , with  $\tau_B = R^2/D_0$ ,  $R$  the radius and  $D_0$  the free diffusion coefficient, or  $Pe_\omega$  if the short-time self diffusion coefficient  $D_s(\varphi)$  is used. At low  $Pe_\omega^0$  yielding is related to Brownian-assisted irreversible particle motion manifested in a Dynamic Strain Sweep (DSS) with the peak of  $G_1''$ . However, at the largely unexplored regime of high  $Pe_\omega^0$ , we detect collision-dominated yielding and, within the oscillation period, a strongly anisotropic structure causing a reduced stress beyond strain reversal (memory of structure due to lack of Brownian relaxation). At intermediate  $Pe_\omega^0$ , the sample is affected by both mechanisms as manifested by a novel, for HS glasses, double peak in  $G_1''$ .

We used sterically stabilized poly(methyl methacrylate) (PMMA) model nearly hard-sphere particles with two radii,  $R = 358$  nm and 130 nm (polydispersity 10–12%) suspended in octadecene and octadecene/bromonaphthalene mixture respectively, in order to expand the  $Pe_\omega$  range. We prepared different volume fractions,  $\varphi$ , by diluting a random close packed batch with the exact  $\varphi$  then adjusted by matching  $G_1'$  of the two systems in agreement with [19]. Experiments were performed on an ARES strain controlled rheometer (with 25mm diameter/0.01rad angle cone-plate) and a solvent trap to eliminate evaporation. In BD simulations, HS interactions were implemented through the potential free algorithm[20]. Oscillatory shear was applied with periodic boundary conditions using typically 5405 particles with 10% polydispersity to avoid crystallization.

In figure 1 we show DSS tests performed at low and high  $Pe_\omega$ , together with the evolution of the normalized intensity of all higher stress harmonics,  $I_{all}/I_1 = \sum I_i/I_1$ , ( $i = 2n + 1$ ,  $n \geq 1$ ), as well as Lissajous curves (intracycle stress versus strain) for representative strain amplitudes,  $\gamma_0$ . The low  $Pe_\omega$  ( $= 0.5$ ) data (fig. 1a) exhibit the typical DSS response with a  $G_1''$  peak around  $\gamma_y$  (at  $G_1' = G_1''$ ) similar to previous studies[11, 19, 21]. The non-linear response is accompanied by progressively larger intracycle non-linearities as indicated both by the Lissajous curves and the increasing  $I_{all}/I_1$  as expected[10, 19]. The latter increases beyond yielding and reaches almost 30% at high  $\gamma_0$  as found previously[19]. Moreover, the Lissajous plots show a transition from a linear viscoelastic behavior (elliptical shape) at low

FIG. 1: Top: Dynamic strain sweeps for (a)  $R = 130$  nm,  $\varphi = 0.60$  at  $\omega = 1$  rad/s ( $Pe_\omega^0 = 0.04$ ,  $Pe_\omega = 0.5$ ) and (b)  $R = 358$  nm  $\varphi = 0.60$  at  $\omega = 1$  rad/s ( $Pe_\omega^0 = 0.9$ ,  $Pe_\omega = 11.2$ ), with the 1st harmonic of the elastic  $G'_1$  and viscous  $G''_1$  modulus as a function of strain amplitude. Middle: Representative Lissajous plots are shown in different strain amplitudes as indicated. Bottom: Normalized total intensity of the higher harmonics of the stress,  $I_{all}/I_1$ .

$\gamma_0$ , to a parallelogram pattern indicative of an intracycle sequence of elastic-plastic response at  $\gamma_0 > \gamma_y$  [10, 19].

At high  $Pe_\omega$ , achieved with large particles ( $R = 358$  nm) at  $\omega = 1$  rad/s ( $Pe_\omega = 11.2$ ) the response is qualitatively different (fig. 1b). Firstly, the peak of  $G''_1$  shifts to higher  $\gamma_0$ , beyond  $\gamma_y$ . Secondly,  $I_{all}/I_1$  (and individual  $I_{2n+1}/I_1$ ) exhibits a non-monotonic behavior showing a first maximum around  $\gamma_y$  and subsequently decreases substantially well inside the non-linear regime. Hence, the sample exhibits a more harmonic stress response (anharmonicity is lowered) even though under non-linear LAOS.

We further explore the  $Pe_\omega$  dependence by changing  $\omega$  while keeping  $\gamma_0$  constant. In fig. 2a experiments with varying  $\omega$  (at  $\gamma_0 = 100\%$ ) reveal the transition from the low  $Pe_\omega$  rectangular shaped Lissajous curves reflecting a sequence of elastic and plastic responses, to the high  $Pe_\omega$  regime with a characteristic *ellipsoid with a double concave distortion* caused by reduced stress in the quadrants II and IV after strain reversal. The intensity of the 3rd

harmonic at 100% (above  $\gamma_y$ ) exhibits a minimum with  $\omega$  in experiments,  $\varphi = 0.62$ , and BD,  $\varphi = 0.60$ , (fig. 2c); note that the position of minimum is  $\varphi$  and  $\gamma_0$  dependent. While in both  $Pe_\omega$  regimes the stress response is highly anharmonic, with significant  $I_3/I_1$ , during the transition the Lissajous curves (fig. 2a) acquire an ellipsoid shape involving almost zero higher harmonic contributions. BD simulations showing identical rheological response (fig. 2b) with experiments are able to provide valuable structural information revealing the underlying mechanism of such stress reduction. In fig. 2d we plot the 2D projection of the pair correlation function in the velocity-gradient (xy) direction,  $g_{xy}(r)$ , at specific points inside the oscillation cycle for  $Pe_\omega = 100$ , similar to findings under steady shear[7]. Contrary to what we find at low  $Pe_\omega$  (supplemental material), here the structure is highly anisotropic at the point of maximum strain (zero shear rate; point A in fig. 2b). Moreover, such anisotropy is persistent during a large part of the successive quadrant where the shear has been reversed (points B and C). Such an anisotropic cage, created during high  $Pe_\omega$  shear in one direction, allows flow with less stress (due to fewer particle collisions) when shear is reversed. Thus, in quadrants II and IV the stress is reduced if compared to a fully harmonic viscous response corresponding to the flowing anisotropic structure of quadrants I and III (dash-dot line in fig. 2b). Only beyond zero strain (points D and E) is the structure reversed and the stress comes back to the maximum values within the period (see supplemental material). Such response is absent at low  $Pe_\omega$  since Brownian motion relaxes shear-induced structural anisotropy more efficiently, and the stress response is similar in all quadrants.

The data presented above verify the existence of the two  $Pe_\omega$  regimes: The low  $Pe_\omega$  one, conventionally studied up to now, where Brownian motion is dominant and the high  $Pe_\omega$  where shear-induced particle collisions introduce novel LAOS features related to the persistent structural anisotropy and the consequent reduced stress after strain reversal. The transition from Brownian activated yielding, where particles under shear escape their cages assisted by thermal motion, to collision-induced cage breaking at high frequencies is linked to pronounced irreversible particle rearrangements and decreasing  $\gamma_y$  at low frequencies[8, 9].

We further investigated HS glasses at higher  $\varphi$  and frequencies corresponding to an intermediate ( $\varphi$  dependent)  $Pe_\omega$  regime. Experimental DSS, Lissajous plots and  $I_{all}/I_1$  shown in fig. 3 as a function of  $\gamma_0$  for  $\varphi = 0.639$  reveal an even richer mechanical response at  $Pe_\omega^0 = 0.4$  ( $Pe_\omega = 8$ ). A main observation here is the unambiguous detection of two peaks

FIG. 2: Lissajous plots at 100% strain amplitude (a) for experiments at different  $Pe_\omega$  denoted by the dash lines in (c) with the 4 quadrants of the oscillatory cycle indicated and (b) BD simulations at  $Pe_\omega = 100$ . The dashed line in (b) indicates a viscous harmonic stress strain response (see also supplemental material). (c)  $Pe_\omega$  dependence of the 3rd harmonic at 100%, for experiments at  $\varphi = 0.62$  ( $R = 358$  nm) and BD simulations at  $\varphi = 0.60$ . (d) 2D projections in the velocity-gradient (xy) plane of the difference of  $g_{xy}(r)$  under shear from that at rest from BD simulations at indicated points within the cycle, points A-E in (b).

in  $G_1''$  detected for the first time in HS glasses. Such a feature was so far observed only in attractive glasses and gels indicating a two-step yielding due to two length-scales present in attractive systems i.e. the interparticle bond and the cage or cluster size[21, 22]. However, the double  $G_1''$  peak seen here must be of a different nature since a second length-scale is absent and moreover, the phenomenon is only observed in a narrow range of  $Pe_\omega$ . The first peak of  $G_1''$  is identified with the one observed at low  $Pe_\omega$  by a direct comparison of the two DSSs ( $Pe_\omega^0 = 0.04$  and 0.4) shown in fig 3a. The two peaks signify the maximum in energy

FIG. 3: (a) Dynamic strain sweeps with  $R = 130$  nm particles at  $\varphi = 0.639$  and an intermediate ( $\omega = 10$  rad/s,  $Pe_\omega^0 = 0.4$ ,  $Pe_\omega = 8$  thick black lines) and low ( $\omega = 1$  rad/s,  $Pe_\omega^0 = 0.04$ ,  $Pe_\omega = 0.8$  thin red lines) frequency regime. (b) Indicative Lissajous plots for  $Pe_\omega = 8$  at different strain amplitudes and (c) Normalized total intensity of higher harmonics  $I_{all}/I_1$ , versus strain amplitude for  $Pe_\omega = 8$ .

dissipation during the two yielding mechanisms at low and high  $Pe_\omega^0$ , attributed to cage breaking via shear-assisted activated hopping and through particle collisions, respectively. The Lissajous figures and higher harmonics reveal a transition from the low  $\gamma_0$  linear response to a viscoplastic flow at high  $\gamma_0$  passing through two states with apparent harmonic response as indicated by the two minima of  $I_{all}/I_1$ . At these strain amplitudes the Lissajous

FIG. 4: BD simulations at  $\varphi = 0.60$ : (a) DSS data showing  $G'_1$  and  $G''_1$  as a function of  $\gamma_0$  for different  $Pe_\omega$  as indicated. (b) Power law exponent for the  $\gamma_0$  dependence of  $G''_1$  and  $D_{eff}(t = T)$  as a function of  $Pe_\omega$  (c) Average  $D_{eff}(t = T)$  versus  $\gamma_0$  for different  $Pe_\omega$  and (d)  $D_{eff}(t = T)$  versus  $Pe_\omega$  for  $\gamma_0 = 10\%$ ,  $30\%$  and  $100\%$ . The corresponding  $D_{eff}(t = T)$  for a system at rest is also indicated.

curves acquire an nearly ellipsoidal shape due to the compensation of the structural phenomena during the transition from low to high  $Pe_\omega$  (as in fig 2c). Note that for this sample ( $R = 130$  nm,  $\varphi = 0.639$ ) the high  $Pe_\omega$  (fig 1b) was not within the experimental window.

Fig. 4a shows  $G'_1$  and  $G''_1$  from BD LAOS tests for  $\varphi = 0.60$  at different  $Pe_\omega$ . The dependence of  $G'_1$  and  $G''_1$  at high  $\gamma_0$  ( $> \gamma_y$ ) follows a power law decrease  $G'_1(G''_1) \propto \gamma_0^{\nu'} (\gamma_0^{\nu''})$  as detected experimentally [11, 19]. While Maxwell-type models give  $\nu' = 2\nu'' = -2$  and MCT (around the glass transition) predicts lower values but similar  $\nu'/\nu''$  ratio [12, 23] experiments in HS glasses show deviations from such simple dependency [19]. In agreement with experiments (fig. 1b and supplemental material), BD simulations give  $Pe_\omega$  dependent exponents (fig 4a) with  $\nu''$  approaching  $-1$  and  $0$  at low and high  $Pe_\omega$  respectively. Therefore, at high  $Pe_\omega$ , where collision activated out-of-cage particle rearrangements are dominant,  $\gamma_0 G''_1 \simeq \sigma(\dot{\gamma}_{\max})$ , a measure of energy dissipation per unit strain, is proportional to shear rate ( $\gamma_0 \omega$ ) similar to the limiting high shear rate viscosity behavior under steady



shear. On the other hand at low  $Pe_\omega$  a  $G_1'' \propto \gamma_0^{-1}$  dependence corresponds to a steady shear shear thinning response of a HS glass at the yield stress plateau with  $\eta_{eff} \propto \dot{\gamma}^{-1}$ . Note that at this stage, the absence of a double  $G_1''$  peak in BD can not be firmly attributed to the absence of hydrodynamic interactions, since BD at high volume fractions could not be conducted at a similar number of data points as experiments due to computational time restrictions.

Further insight in the two yielding mechanisms is gained by examining microscopic particle dynamics within a LAOS cycle by BD. In Fig. 4c we show the effective diffusivity,  $D_{eff}(t = T) = \langle \Delta z^2(T) \rangle / T$  with  $\langle \Delta z^2(T) \rangle$  the mean square displacement in the vorticity direction,  $z$ , (at  $t = T$ ), as a function of  $\gamma_0$  for several  $Pe_\omega$ . At low  $Pe_\omega$ ,  $D_{eff}(T)$  increases sublinear with  $\gamma_0$ , whereas as  $Pe_\omega$  is increased it exhibits progressively a weaker increase at small  $\gamma_0$  and a stronger one at higher. The constant  $D_{eff}(T)$  at low  $\gamma_0$  indicates that prior to yielding in-cage diffusion is unaffected by shear. For  $\gamma_0 > \gamma_y$ ,  $D_{eff}(T)$ , corresponding to out-of cage diffusion, increases sublinear ( $\gamma_0 = 100\%$ , fig. 4d) with a power law exponent  $\simeq 0.8$  at low  $Pe_\omega$ , which approaches 1 for  $Pe_\omega > 1$  where direct particle collisions are dominant. In comparison, the corresponding  $D_{eff}(T)$  for  $\gamma_0 = 10\%$  and  $30\%$  show an initial sublinear dependence at low  $Pe_\omega$  and subsequently approach the curve at rest, since for such frequencies  $\gamma_y > 30\%$  [9] and the sample has not yet yielded. Note that a similar power law increase (with exponent  $\sim 0.8$ ) has been detected in HS glasses at low steady shear rates by confocal microscopy [6], a weak but systematic deviation from the linear MCT prediction[24] and closer to agreement with non-linear Langevin equation theory involving activated hopping mechanisms [25]. For our oscillatory BD a linear dependence is reached for  $Pe_\omega > 10$  as expected for non-Brownian particles under steady shear[26]. We suggest that the sublinear and linear dependencies probed here at low and high  $Pe_\omega$ , respectively, reflect the two different mechanisms involved in the two regimes. Note that similarly with  $D_{eff}$ ,  $\gamma_0 G_1''$  increases linearly with  $\gamma_0$  at high  $Pe_\omega$ ; the linear  $\gamma_0$  dependence of both quantities is linked to collision-induced yielding. On the other hand at low  $Pe_\omega$  both quantities increase sublinearly,  $D_{eff} \propto \gamma_0^{0.8}$  and  $\gamma_0 G_1'' \propto \gamma_0^{0.4}$  (the latter exponent tends to zero as  $Pe_\omega$  is lowered) reflecting plastic flow, with a power law stress behavior, and complex Brownian/shear-activated particle hopping.

In summary, the combination of experimental oscillatory rheology and BD simulations has revealed the complete mechanical fingerprint of HS glasses and the related underlying

microscopic structure and dynamics over a wide frequency regime where both Brownian and non-Brownian behavior is probed. At low  $Pe_\omega$ , commonly studied up to now, Brownian-assisted irreversible motion takes place during yielding with a single peak of  $G_1''$  and strong higher harmonics of the stress at large  $\gamma_0$ . In this regime the microstructure under shear is only weakly anisotropic while the shear-induced diffusivity scales sublinear with  $Pe_\omega$ . At high  $Pe_\omega$  yielding is dictated by collision-induced displacements linked with a shear-induced long time diffusion that increases linearly with  $Pe_\omega$ . A single  $G_1''$  peak is detected at  $\gamma_0 > \gamma_y$  which eventually turns into a plateau at the limit of large  $Pe_\omega$ . The structure under shear is strongly anisotropic but more interestingly exhibits a hysteresis under strain reversal. Such structural memory due to lack of Brownian relaxation, causes a characteristic stress drop after strain reversal as revealed by the Lissajous curves. At some characteristic  $\gamma_0$ , higher harmonics drop to almost zero indicating an unexpected harmonic response even though the sample is under non-linear shear. Finally, at a  $\varphi$  dependent intermediate  $Pe_\omega$  regime the sample is affected by both mechanisms (Brownian- and collision-induced yielding) resulting in a double peak of  $G_1''$  and two minima of higher harmonics, a feature that is detected for the first time in simple hard sphere glasses.

The rich mechanical response of a model hard sphere glass revealed here in oscillatory shear as a function of frequency, bridging the Brownian and non-Brownian regimes, may provide insights for the understanding of systems with more complicated interparticle interactions such as pastes, slurries, particle gels, jammed emulsions and metallic glasses under a wide range of conditions.

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